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Determination of ^{99}Mo, ^{103}Ru, and ^{140}Ba Activity in an Irradiated Solution of Low Enriched Uranium Using Gamma Ray Spectroscopy

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ABSTRACT

The need for technologies and methods to produce ^{99m}Tc outside of the conventional nuclear reactor process has continued to grow over the past decade. Consequently, more work and research have been done to produce ^{99}Mo using less traditional methods. In this work, a 300.7 mL solution containing low enriched uranium was irradiated with a linear accelerator. A portion of the sample was then counted with a gamma ray spectrometer to determine the activities of ^{99}Mo, ^{103}Ru, and ^{140}Ba in the solution at the end of the irradiation. The activities at the end of the irradiation were determined to be 68.00 ± 3.84, 3.835 ± 0.226, and 20.47 ± 1.07 mCi, respectively, in the 300.7 mL solution.

INTRODUCTION

As technetium-99m (^{99m}Tc) is one of the most widely used diagnostic imaging agents, the reliability of its supply is paramount in the medical radioisotope production industry [1]. An estimated 3-5% annual increase in demand for ^{99m}Tc compounded with the recent shutdown of the National Research Universal reactor at Chalk River in Ontario, Canada has led to the subsequent shortage of its parent isotope, molybdenum-99 (^{99}Mo), jeopardizing the supply of ^{99m}Tc to the medical industry [1-3].

Almost all of the ^{99}Mo supplied to the medical industry is being produced via the uranium-235 (^{235}U) fission route using highly enriched uranium (HEU) targets in nuclear reactors. Following irradiation, ^{99}Mo is separated from the HEU target and allowed to decay to its daughter for use in patients. Because HEU poses a higher proliferation concern, support for it, in any form, has decreased, while the demand for ^{99m}Tc continues to increase. Therefore, new methods and technologies for producing ^{99}Mo have gained more attention and research [1].

This report is concerned with the method of bombarding a low enriched uranium (LEU) solution target with neutrons from a linear accelerator. This still utilizes the ^{235}U fission route; however, the goal is to effectively produce ^{99}Mo using LEU, rather than HEU. For this work, a 300.7 ± 0.1 mL liquid solution containing 45.11 ± 0.01 g of dissolved U enriched to 19.54% ^{235}U was irradiated at the LANSCE facility at the Los Alamos National Laboratory. The purpose of this irradiation was to produce and extract a quantity of ^{99}Mo to further the research in replacing HEU targets with LEU solution targets while continuing to supply the demand for ^{99m}Tc. After a decay period of a few days, a 12.27 ± 0.01 mL sample was extracted from the irradiated solution and placed in a vial for analysis using gamma ray spectroscopy.

DESCRIPTION OF THE WORK

Dimensions

The 12.27 mL sample was placed in a 20.83 mL vial for counting. The thickness of the vial wall was 0.127 cm. The sample was placed inside a hot cell to reduce the dose to personnel. The gamma ray spectrum was taken through a 9.53 cm diameter port hole in the 81.28 cm thick wall using an Ortec Transpec gamma ray spectrometer with a 5.08 cm lead (Pb) collimator. The collimator was lined with copper to reduce interference from fluorescent Pb X-rays. The detector has a relative efficiency of 45%, and was placed 214.24 ± 0.01 cm from the sample. A digital range finder was used to determine this distance. Though the liquid source was cylindrical in shape due to the vial in which it was placed, it was of sufficient distance from the detector to be considered a point source. The solid angle subtended by the detector was determined to be $(7.545 \pm 0.101)\times10^{-4}$ sr.

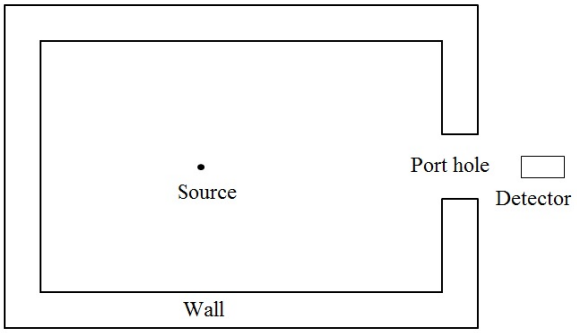

Figure 1. A sketch of the experimental setup, not drawn to scale.

Detector Efficiency

The intrinsic efficiency of the detector was previously determined, and the geometry-corrected efficiency was calculated by multiplying the intrinsic efficiency by the solid angle.

Counting

The sample was counted with the detector in this geometry for a live time of 1300 seconds. The dead time was 37.66%, leading to a real time of 2085.34 s.

Calculating Activity

From the number of background-subtracted counts gathered at each gamma ray peak for the nuclides of interest in the irradiated spectrum, the activities at the end of the irradiation of the associated isotope can be determined using Equation 1.

$$A = \frac{Counts}{t_L I \varepsilon T e^{-\lambda t_R} e^{-\lambda t_d}} \qquad (1)$$

where A is the calculated activity at the end of irradiation, *Counts* is the measured number of counts under the energy peak of interest, t_L is the live time of the detector, I is the gamma ray emission intensity for the measured energy, ε is the efficiency of the detector at the measured energy, T is the transmission fraction of gamma rays at the measured energy through the sample material, λ is the decay constant of the nuclide being measured, t_R is the real time of the count, and t_d is the decay time between the end of irradiation and the beginning of the count. The transmission fractions were estimated using a water solution with a density of 1.192 g/cm^3. The emission intensities and their associated error are given in the literature [3]. There is an assumed 5% error due to the estimation of these values. It is important to note that the activity calculated using Equation 1 will be that of the measured sample and must be corrected for the total volume of the irradiated solution.

RESULTS

The measurements were carried out on three nuclides of interest. First and foremost is ^{99}Mo at the 181.07, 739.50, and 777.92 keV peaks. The activities of ruthenium-103 (^{103}Ru) and barium-140 (^{140}Ba) were also calculated using the 497.08 and 610.33 keV peaks for ^{103}Ru and the 537.26 keV peak for ^{140}Ba [3]. Table 1 shows the intensities, efficiencies, and transmission fractions at these energies. Table 2 gives the measured counts and the calculated activities of each nuclide at their respective gamma ray energy in the total 300.7 mL irradiated solution.

Table 1. Intensities (I), efficiencies (ε), and transmission fractions (T) for the nuclides of interest

Nuclide	Energy (keV)	I	ε (×10^{-5})	T
^{99}Mo	181.07 ± 0.01	0.0599 ± 0.0011	3.349 ± 0.048	0.8536 ± 0.0427
^{99}Mo	739.50 ± 0.02	0.1213 ± 0.0022	1.290 ± 0.017	0.9046 ± 0.0452
^{99}Mo	777.92 ± 0.02	0.0426 ± 0.0008	1.240 ± 0.017	0.9067 ± 0.0453
^{103}Ru	497.08 ± 0.01	0.9100 ± 0.0122	1.793 ± 0.024	0.8881 ± 0.0444
^{103}Ru	610.33 ± 0.02	0.0576 ± 0.0006	1.509 ± 0.020	0.8967 ± 0.0448
^{140}Ba	537.26 ± 0.01	0.2439 ± 0.0007	1.680 ± 0.022	0.8914 ± 0.0446

Table 2. Counts measured and calculated activities of the nuclides of interest

Nuclide	Energy (keV)	Counts	A (mCi)
^{99}Mo	181.07 ± 0.01	62,116 ± 450	67.60 ± 3.75
^{99}Mo	739.50 ± 0.02	52,286 ± 266	67.74 ± 3.73
^{99}Mo	777.92 ± 0.02	17,917 ± 179	68.65 ± 4.03
^{103}Ru	497.08 ± 0.01	95,118 ± 382	3.665 ± 0.190
^{103}Ru	610.33 ± 0.02	5594 ± 195	4.005 ± 0.261
^{140}Ba	537.26 ± 0.01	111,136 ± 419	20.47 ± 1.07

The activities calculated for each nuclide at their respective gamma ray energy peaks agree very well with one another.

CONCLUSION

A 300.7 mL solution containing 45.11 g of LEU enriched to 19.54% ^{235}U was irradiated using the linear accelerator at LANSCE. After allowing the solution to decay for a few days, the activities of ^{99}Mo, ^{103}Ru, and ^{140}Ba were measured using gamma ray spectroscopy. The average of the measured activities for each nuclide were 68.00 ± 3.84, 3.835 ± 0.226, and 20.47 ± 1.07 mCi, respectively, in the 300.7 mL solution.

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